used. Other species concentrations at the boundaries, as well as all species at the top of the modeling domain, are set to tropospheric clean-air concentrations.

Meteorological data are assimilated by the first stage of preprocessors. These data contain regular hourly observations from U.S. National Weather Service surface stations (and from similar stations in Canada as necessary), including wind speed and direction, air temperature, dew point, atmospheric pressure, and cloud amounts and heights. Twice-daily sounding data, from the upper-air observation network also are included in the meteorological database. Upper-air meteorological parameters include atmospheric pressure, wind speed and direction, and air temperature, dew point. Finally, both buoy and Coastal Marine Automated Station data are used. The parameters that typically are reported include wind speed and direction and air and sea temperatures.

Emissions data for the primary species are input to the ROM system as well. Originally these data were provided from the 1985 emissions inventory of NAPAP, with 18.5-km spatial resolution. Most recently, the interim regional inventory has been used widely to support current applications of the ROM. It represents an update and improvement of the NAPAP inventory and is being used to support SIP modeling until state inventories are approved (U.S. Environmental Protection Agency, 1993a,b). Species included are CO, NO, NO_2, and 10 hydrocarbon reactivity categories. Natural hydrocarbons also are input, including isoprene explicitly, monoterpenes divided among the existing reactivity classes, and unidentified hydrocarbons. The chemical mechanism in ROM is the CBM-IV.

Land-use input data consist of 11 land-use categories in 1/4□ longitude by 1/6□ latitude grid cells. The data are more than 20 years old and represent a weakness. New land-use data slowly are being collected and released. Changes in land use over the last 20 years may change significantly the estimates of biogenic hydrocarbon emissions for large regions of the United States. Data are provided for the United States and Canada as far as 55□ N. The land-use categories are (1) urban land, (2) agricultural land, (3) range land, (4) deciduous forests, (5) coniferous forests, (6) mixed-forest wetlands, (7) water, (8) barren land, (9) nonforested wetland, (10) mixed agricultural land and range land, and (11) rocky, open places occupied by low shrubs and lichens. Land-use data are used to obtain biogenic emissions estimates, as a function of the area of vegetative land cover, and for the determination of surface heat fluxes.

Topography input data consist of altitude matrices of elevations in a 7.5□ × 7.5□ grid. The data are obtained from the GRIDS database operated by EPA's Office of Information Resources Management. Topography data are used in the calculation of layer heights.

The ROM does have its limitations, including the large grid size, relatively crude wind fields, and highly empirical vertical mixing assumptions (Wolff, 1993).

3.6.3.3 The Regional Acid Deposition Model

The RADM initially was developed at the NCAR for EPA and subsequently was refined and improved at the State University of New York at Albany. The model is an Eulerian transport, transformation, and removal model that includes a treatment of the relevant physical and chemical processes leading to acid deposition and the formation of photochemical oxidants. As summarized in Tables 3-21 through 3-25, these processes include atmospheric transport and mixing, gas-phase and aqueous-phase chemical transformations, dry deposition, and cloud mixing and scavenging.

Chemical trace species are transported and diffused through the three-dimensional RADM grid using externally specified meteorological data. The RADM uses hourly three-dimensional fields of horizontal winds, temperature, and water vapor mixing ratio calculated by the meteorological model MM4 with FDDA. In addition, RADM requires two-dimensional, hourly fields of surface temperature, surface pressure, and precipitation rates over the model domain. Kuo et al. (1985) found that in order to calculate accurate mesoscale trajectories, at least 3-h temporal resolution is desirable, and the 12-h resolution of upper air observations is inadequate. Recent verification studies with 30 meteorological episodes by Stauffer and Seaman (1990) further support the use of MM5 data with FDDA. Using meteorology generated from a dynamically consistent meteorological model can introduce errors caused by simulation errors associated with the meteorological model. These uncertainties can be quantified through objective verification studies with observed data (Anthes et al., 1985; Stauffer and Seaman, 1990).

The RADM2 chemical mechanism has been described by Stockwell et al. (1990), Chang et al. (1991b), Carter and Lurmann (1990), and Stockwell and Lurmann (1989). For RADM2, the VOCs are aggregated into 12 classes of reactive organic species. Each category of VOC is represented by several model species that span the required range for reaction with the OH radical. Most emitted organic compounds are lumped into surrogate species of similar reactivity and molecular weight, although organic chemicals with large emissions are treated as separate model species even though their reactivities may be similar. Categories of VOCs with large reactivity differences and complicated secondary chemistries are represented by larger numbers of intermediate and stable species. During the aggregation of organic species, the principle of reactivity weighting is followed to attempt to account for differences in reactivity.

A major part of the SARMAP program described earlier is the extension of the RADM. The SARMAP is the modeling and data analysis component of a multi-year collaboration between two projects— SJVAQS and AUSPEX. In the near term, the objective of SARMAP is to produce a model that can be used to examine scenarios for control of O_3 precursor emissions as required under the CAAA for the 1994 planning cycle. The goals of the SARMAP modeling program can be summarized as follows:

- Development of a comprehensive state-of-the-science three-dimensional modeling system (consisting of emissions, meteorological, and air quality models) suitable for the simulation of O_3 concentrations, PM_{10} concentrations, visibility degradation, and acid deposition;
- Evaluation of the modeling system and its individual components against experimental data collected during the SJVAQS/AUSPEX field program; and
- Application of the model to estimate the effect of changes in emission levels on O_3 concentrations, PM_{10} concentrations, visibility degradation, and acid deposition.

The general attributes of the SARMAP modeling system are listed below.

- Integrated system of individual modules, including air quality, meteorological, emissions, and emissions projection; full compatibility of gridding system among all models.
- Ozone estimation capability; capability for efficiently incorporating modules for simulating aerosols, visibility, and acid deposition.
- Applicability at urban, subregional, and regional scales, embodying a full range of anticipated physical, chemical, and terrain characteristics.

- Capability of being driven by larger meteorological models, if desired (for generating initial and boundary conditions).
- Capability of generating as output a full complement of chemical species concentrations and meteorological parameters.
- Variable horizontal grid size.
- Variable number of vertical layers.
- Variable depth of vertical layers.
- Capability of nested grid application.
- Capability of varying the number of vertical layers with time of day. Selection of number of layers and timing of changes to be model-driven.
- Improved treatment of emissions injection aloft, including placement of plumes in the vertical, treatment of inversion penetration, proper vertical dilution of plumes, and proper treatment of chemistry.
- Inclusion of plume-in-grid capability.
- Capability for use of "computational tracers" for a variety of tests.
- Capability of simulating the O_3-VOC-NO_x system alone or in tandem with the aerosol system.
- Capability of simulating aerosols for the O_3-VOC-NO_x system.

The following modifications to the RADM2 gas-phase chemical mechanism have been made:

- Updating the rate constants, product parameters, and absorption cross sections and quantum yields for consistency with current recommendations;
- Improving the treatment of isoprene chemistry;
- Adapting the SAPRC emissions processing scheme to the RADM2 mechanism; and
- Adding extra species (acetaldehyde, PAN, and an additional aromatic) and their associated reactions and products.

The Smolarkiewicz scheme currently used in RADM will be replaced with the Bott scheme. This scheme is more accurate than the Smolarkiewicz scheme for continuous plumes and at low grid resolutions. The RADM cloud module will be replaced with the ADOM module. The RADM dry deposition module currently underestimates dry deposition velocities under stable conditions. This can result in unrealistically high O_3 concentrations at night.

3.6.4 Evaluation of Model Performance

Air quality models are evaluated by comparing their predictions with ambient observations. Because a model's demonstration of attainment of the O_3 NAAQS is based on hypothetical reductions of emissions from a base-year-episode simulation, the accuracy of the base-year simulation is necessary, but not sufficient. An adequate model should give accurate predictions of current peak O_3 concentrations and temporal and spatial O_3 patterns. It should also respond accurately to changes in VOC and NO_x emissions, to differences in VOC reactivity, and to spatial and temporal changes in emissions patterns for future years.

Model performance can be evaluated at several levels. The important sub-models, the emissions model, the meteorological model, and the chemical mechanism can be evaluated independently, and the model as a whole can be evaluated. Evaluation of emissions models can be carried out with special measurements designed to isolate the effects of emissions from a particular source category, such as tunnel studies (Pierson et al., 1990) or on-road

surveillance of motor vehicles (Lawson et al., 1990) to evaluate the accuracy of motor vehicle emissions models. Meteorological sub-models can be evaluated from the results of tracer experiments. Chemical mechanisms have traditionally been developed and evaluated on the basis of smog chamber experiments. A question that merits continued attention is how well chemical mechanisms developed with reference to smog chamber data perform when simulating the ambient atmosphere. As noted in this section, comparisons of observed and predicted concentrations for all important precursors, intermediates, and products are important in assessing the accuracy of a chemical mechanism.

Compilations of the performance of photochemical models in the South Coast Air Basin of California and in other urban areas indicate a general tendency toward the underprediction of O_3 concentrations and particularly O_3 maxima. It should be noted that different areas of the country are characterized by different controlling factors in O_3 generation, so the reasons for O_3 underprediction in one area may not be the same as in another. A case in point is the possibility of anthropogenic ROG emissions underestimation in urban areas versus biogenic ROG emissions underestimation in rural and regional areas. It is well-recognized that urban and regional photochemical models have a number of uncertain input quantities, so it is possible, by adjusting these quantities within their ranges of uncertainty, to improve O_3 predictions. This process, which is inherent in any modeling exercise because of the uncertainty associated with many of the input quantities, can lead to getting the right answer for the wrong reason. Because the modeling of an O_3 episode usually is carried out to establish a "base case" against which to evaluate the effects of VOC and NO_x emissions changes, the accuracy of the base case is vital for obtaining a valid assessment of the effects of emissions perturbations. Due to the nonlinear response of the O_3/VOC/NO_x system, conclusions drawn about the effect of VOC and NO_x emissions changes may not reflect actual atmospheric response if the base case simulation is inaccurate. For this reason, it is important to understand the reasons why the base case simulation may not agree with observations. Several more or less equivalent alternate base cases may exist due to the fact that it often is possible to vary inputs within their ranges of uncertainties to achieve comparable model performance. Unfortunately, the O_3 responses to identical VOC/NO_x controls may be rather different depending on which base case is used.

3.6.4.1 Model Performance Evaluation Procedures

Specific numerical and graphic procedures have been recommended for evaluation of the accuracy of grid-based photochemical models (Tesche et al., 1990b). The recommended methods include the calculation of peak prediction accuracy; various statistics based on concentration residuals; and time series of predicted and observed hourly concentrations. Four numerical measures appear to be most helpful in making an initial assessment of the adequacy of a photochemical simulation (Tesche et al., 1990b): (1) the paired peak prediction accuracy, (2) the unpaired peak prediction accuracy, (3) the mean normalized bias, and (4) the mean absolute normalized gross error.

Accurate matching of O_3 alone may not be sufficient to ensure that a model is performing accurately. The possibility of compensatory errors must be recognized (in which two or more sources of error interact in such a way that O_3 is predicted accurately, but for the wrong reasons). The inaccuracies offset each other in part. The modeling effort should be designed to minimize the likelihood of the presence of compensatory errors.

Evaluation of model performance for precursor and intermediate species, as well as for product species other than O_3, when ambient concentration data for these species are available, significantly improves the chances that a flawed model will be identified. Comparisons of observed and predicted concentrations for all important precursors, intermediates, and products involved in photochemical air pollution, such as individual VOCs, NO, NO_2, PAN, O_2, H_2O_2, HNO_2, and HNO_3, are useful in model evaluation, especially with respect to the chemistry component of the model (Jeffries et al., 1992). Comparisons of predictions and observations for total organic nitrates (mainly PAN) and inorganic nitrates (HNO_3 and nitrate aerosol) can be used to test qualitatively whether the emissions inventory has the correct relative amounts of VOCs and NO_x. However, in order to include HNO_3 and nitrate aerosol in the data set for model comparisons, the model should include an adequate description of the HNO_3 depletion process associated with aerosol formation.

Adequate model performance for several reactive species increases the assurance that correct O_3 predictions are not a result of chance or fortuitous cancellation of errors introduced by various assumptions. Multispecies comparisons could be the key in discriminating among alternative modeling approaches that provide similar predictions of O_3 concentrations.

As noted above, photochemical models have the potential to produce nearly the right O_3 concentrations when performance is evaluated, but do so because two or more flaws were compensating each other. The existence of compensating errors in many modeling applications is suspected because most applications have used emission inventories whose validity is now in question (National Research Council, 1991). Underestimation of VOC emissions from motor vehicles may be responsible for the lack of agreement between inventories and ambient concentration data (Baugues, 1986; Lawson et al., 1990; Pierson et al., 1990; Fujita et al., 1992). Underestimation of emissions from other sources is also a possibility. One potentially underestimated VOC source is vegetation, which naturally emits VOCs. An underestimation of VOC emissions could be compensated for by underestimation of mixing height or wind speed, by overestimation of boundary concentrations of O_3 or precursors, or by inaccurate chemistry modules. Boundary concentrations (which can be obtained from measurements or regional models or by assuming background concentrations, often are poorly defined.

If only a routine database is available for modeling O_3 in an urban area, then there are four areas of concern that require attention (Roth, 1992).

(1) Air Quality Aloft. These data most likely will not be available. These measurements are important and are instrumental for diagnostic analysis of model simulations.

(2) Boundary Conditions. If the possibility of significant transport into the region exists, but the data are not available, the boundary conditions become a variable that allows the introduction of compensatory errors if the emissions estimates are inaccurate. An approach to circumventing this problem is to define the region in such a way that the boundaries become a much less significant issue.

(3) Ambient VOC Data. These generally are not routinely available. In their absence, evaluation of model performance is hampered.

(4) Meteorological Data Aloft. Very often, there are only surface measurements and a few soundings from which to extrapolate the needed data.

If any of these four areas is missing from the database, the performance evaluation and subsequent model application must be planned to minimize the possibility of compensatory errors.

3.6.4.2 Performance Evaluation of Ozone Air Quality Models
Urban Airshed Model

The UAM has been applied to many urban areas in the United States and Europe, and most of these studies have included some form of performance evaluation (see summary in Tesche et al., 1993, Table 6-2). Thus, there is a growing body of information concerning the accuracy of the model's predictions; UAM itself is continuing to undergo revision. Evaluations of UAM's performance have been carried out for a number of geographic areas. Evaluations conducted since 1985 have indicated mean discrepancies between predicted and measured O_3 values of 20 to 40% of the observations, when paired in space and time (Roth et al., 1990). The prediction of peaks exhibits relative errors that are smaller than the average error, with a tendency toward underprediction (Roth et al., 1990). The discrepancies between predicted and measured NO_2 in UAM applications are on the order of 30 to 50%, with no improvement over the history of modeling applications (Roth et al., 1990). Underprediction of NO_2 by UAM has been typical, generally on the order of 20 to 40% (Roth et al., 1990).

As a result of the discovery of significantly underestimated mobile source VOC emissions (in the late 1980s), this emissions underestimation is the leading cause of O_3 underprediction in urban areas.

Regional Oxidant Model

A primary role of the ROM is to estimate boundary conditions for use by UAM in evaluating hydrocarbon and NO_x reduction strategies for urban areas in the eastern United States. This is especially the case in areas where transport is a significant element (U.S. Environmental Protection Agency, 1990d). Analysis of regional O_3 abatement strategies also is a major role of the ROM (Possiel et al., 1990).

The ROM has been used in the EPA program, the Regional Ozone Modeling for Northeast Transport (ROMNET) program, to assess the effectiveness of various regional emission control strategies in lowering O_3 concentrations to nationally mandated levels for the protection of human health, forests, and crops (Meyer et al., 1991b). As part of the ROMNET program, the ROM also is being used to provide regionally consistent initial and upwind boundary conditions to smaller-scale urban models for simulations of future-year scenarios.

The most complete testing of ROM2.0 was accomplished in an evaluation with the 50-day (July 12 to August 31, 1980) Northeastern Regional Oxidant Study database (Schere and Wayland, 1989a,b). The model underestimated the highest values and overestimated the lowest. It produced an overall 2% overprediction in predicting maximum daily O_3 concentrations averaged over aggregate groups of monitoring stations. A key indicator of model performance on the regional scale is the accuracy of simulating the spatial extent and location, as well as the magnitude, of the pollutant concentrations within plumes from significant source areas. In ROM2.0 performance analyses, plumes from the major metropolitan areas of the Northeast Corridor, including Washington, DC; Baltimore, MD; New York; and Boston, could be clearly discerned in the model predictions under episodic conditions. Generally, the plumes were well characterized by the model, although there was evidence of a westerly transport bias and underprediction of O_3 concentrations near the center

of the plume. Using aircraft data, ROM2.0 was found to underpredict the regional tropospheric burden of O_3.

The evaluation of ROM2.1 (Pierce et al., 1990), unlike that of ROM2.0, was based on routinely archived data from state and local agency monitoring sites rather than on an intensive field-study period. The evaluation consisted of the comparison of observed and predicted O_3 concentrations during selected episodes (totaling 26 days) of high O_3 observed during the summer of 1985. Evaluation showed that ROM2.1 underestimated the highest values and slightly overestimated the lowest; underestimates of the upper percentiles tended to be more prevalent in the southern and western areas of the ROMNET domain (Table 3-26). The model exhibited an overall 1.4% overprediction in predicting maximum daily O_3 concentrations averaged over aggregate groups of monitoring stations, and it appears to correct for the westerly transport bias of high-O_3 plumes in the Northeast Corridor seen in ROM2.0. As with ROM2.0, model performance degraded as a function of increasingly complex mesoscale wind fields.

In a recent evaluation of ROM (Systems Applications International, 1993), ROM2.2 overestimated observed O_3 maxima by 20 to 30 ppb over the period of July 4 through 6, 1988, and predicted an episodic peak of 242 ppb on July 9, 1988, when the observed peak was 138 ppb. The ROM2.2 performance for hourly O_3 concentrations in the New York region exceeded the range of EPA acceptable performance by a factor of two 90% of the time during the July 1988 episode. The Systems Applications International (1993) report concluded that "the patchiness of the ROM2.2 predictions compared to the observations raises serious questions as to whether the model will respond correctly to emission control strategies." The major conclusions of that report were:

- Model performance downwind of New York City is "unacceptable". The model significantly overpredicts peak O_3 levels, and the predicted diurnal variation of O_3 occurs too late in the afternoon.
- Model performance for the Philadelphia and Baltimore/Washington urban plumes is "poor" with "unpaired peak estimation accuracy at the outer edge of the acceptable range."
- Elsewhere, the model seems to give good results, although it produces O_3 spatial distributions that are too "patchy" when compared to observations.
- There is a systematic westerly bias in the ROM2.2 wind fields.
- The model performance for NO_x is "extremely poor" indicating that ROM2.2 may be overestimating the VOC/NO_x ratios across the region.

3.6.4.3 Database Limitations

As previously mentioned, the use of routine air quality and meteorological data requires that a number of assumptions be made about key model inputs. Although intensive field studies are desirable during O_3 episodes to acquire the full set of data required, three key problems arise: (1) such studies are expensive and, therefore, are limited in number; (2) the time required to carry out field studies usually exceeds the time available; and (3) most field studies have not captured the worst O_3 episodes. Because EPA guidance emphasizes planning to meet worst-case conditions, field data often must be manipulated to approximate highest O_3 concentrations. Such adjustments invariably increase uncertainty in model projections.

Studies that have, or will, provide data for model evaluation include the St. Louis, MO, RAPS, conducted in 1975 and 1976; the Northeast Corridor Regional Modeling Project,

conducted in 1979 and 1980; the South Central Coast Cooperative Aerometric Monitoring Program, conducted in 1985; SCAQS conducted in 1987; studies in Sacramento and San Diego, CA, in 1990; SJVAQS/AUSPEX conducted in 1990; LMOS conducted in 1990 and 1991; SOS conducted in 1991 and 1992; and a Gulf Coast study for 1993.

In most cases, field studies have not coincided with periods in which ozone concentrations have attained values as high as that on which the SIP must be based. Given the low probabilities of occurrence of the most adverse meteorological conditions and the fact that field studies typically acquire data for two or three ozone episodes, obtaining a design value concentration during the course of a field study is unlikely.

The EPA recommends that the five highest daily maximum O_3 concentrations at a design-value site, selected from the three most recent years, be modeled if EKMA is used for a SIP (U.S. Environmental Protection Agency, 1989b). Because EKMA's data requirements are minimal, it can be applied to the worst cases. In contrast, the number of episodes available for grid-based modeling is less than desirable in all areas. In addition, any available intensive databases often do not include the worst-case meteorology; intensive databases typically restrict modeling to two or three O_3 episodes having a duration of 2 to 3 days each. Moreover, the intensive databases never encompass the full range of meteorological conditions of interest (if O_3 exceedances occur in an area under different meteorological conditions, the relative effectiveness of different control strategies might vary with the different meteorological conditions). The EPA specifies procedures for episode selection for use with grid-based models (U.S. Environmental Protection Agency, 1991b).

Because the number of intensive databases is limited both in terms of episodes and regions, EPA has investigated the feasibility of applying UAM without conducting intensive field studies (Scheffe and Morris, 1990, 1991). These studies, known as the Practice for Low-cost Application in Nonattainment Regions (PLANR), were conducted for New York; Philadelphia; Atlanta; Dallas-Fort Worth, TX; and St. Louis. Of the five cities studied, St. Louis, New York, and Philadelphia had intensive databases available. Simulations were carried out using both routine and intensive databases for St. Louis and Philadelphia. Model performance using routine data was much better for St. Louis than for Philadelphia (Scheffe and Morris, 1990, 1991). Scheffe and Morris (1990, 1991) cautioned that the differing results may be complicated by the quality of the databases, but they speculate that model performance using routine databases for Philadelphia might have been poorer because of regional transport. Performance statistics for all four applications using routine data were consistent with other UAM applications (Scheffe and Morris, 1990, 1991); however, the paucity of data in the routine databases precluded any investigation of the possibility that compensating errors occurred.

Scheffe and Morris (1990, 1991) note that the PLANR lack of air quality data was addressed by extending the length of the simulations and expanding the upwind boundary, which, in effect, increased the need for accurate emissions inventories (boundary conditions could also be obtained through use of ROM). For PLANR applications, gridded emissions were created from routine county-level emission inventories by utilizing an emissions program that made use of surrogate information, such as population distribution. The PLANR study represents an interesting start on the problem of model application to areas without intensive databases; however the results were not sufficiently definitive for drawing conclusions of a broad, general nature.

3.6.5 Use of Ozone Air Quality Models for Evaluating Control Strategies

Photochemical air quality models are used for control strategy evaluation by first demonstrating that a past episode, or episodes, can be adequately simulated and then reducing hydrocarbon or NO_x emissions in the model inputs and in assessing the effects of these reductions on O_3 in the region. Ozone concentrations can be decreased by reducing either VOC or NO_x concentrations to sufficiently low levels. The effects of NO_x emissions reductions on O_3 concentrations vary because NO_x is an atypical precursor (i.e., although it is necessary for O_3 formation, fresh NO emissions remove O_3, and high concentrations of NO_x retard the rate of O_3 formation by removing radicals). Control of NO_x tends to accelerate the rate of O_3 formation; however, its effects on peak O_3 concentration depend on the location and timing of the control and on ambient concentrations of VOCs and NO_x, which vary widely in time and space, even within a single urban area during 1 day.

At a given VOC level, as the initial NO_x is increased, O_3 first increases, then peaks, and then decreases. The reduction in peak O_3 with increasing NO_x is a well-established chemical phenomenon. The peak in O_3 formation occurs at an initial VOC/NO_x ratio of about 10/1 (i.e., 10 ppbC/1 ppb). At fixed NO_x level, as VOC is increased, O_3 formation increases but then levels off. As a result of this behavior, at VOC/NO_x ratios below about 10/1, VOC reduction has been the preferred strategy for O_3 reduction. In this region NO_x reductions speed up O_3 formation and lead to higher peak O_3 values. At VOC/NO_x ratios exceeding about 10/1, both VOCs and NO_x will reduce O_3, but less than proportionally. The reason the reduction in O_3 is less than proportional is because equal reductions of VOCs and NO_x at intermediate ratios tend to keep O_3 production at its maximum. The nonlinear chemical behavior of the VOC/NO_x system, discussed earlier in this chapter, is at the heart of the controversy over the role of NO_x in O_3 control (Heuss and Wolff, 1993).

As noted in Section 3.6.1.2, the concept that a region is characterized by a single VOC/NO_x ratio is oversimplified and may actually lead to incorrect conclusions concerning the optimal approach to O_3 reduction (Milford et al., 1989). The VOC/NO_x ratio in a region is a function of location and time of day; the source-rich center city area may be characterized by a lower ratio than that in downwind, suburban areas at any given time of day. Because of the complex spatial and temporal dependence of O_3 formation, grid-based photochemical air quality models are necessary to evaluate the effect of emission reduction strategies for a region.

Moreover, location-specific studies need to be performed to ascertain whether a given area is in the VOC- or NO_x-controlled regime. Research is being conducted into the relationship between O_3 and NO_y to determine whether NO_y is a better indicator of the O_3-forming potential than the VOC/NO_x ratio (Shepson et al., 1992b; Trainer et al., 1993; Kleinman et al., 1994; Milford et al., 1994).

In most modeling applications, inputs are adjusted within their range of uncertainty to improve performance. A key test of quality of performance is to evaluate the model predictions for other episodes without adjustments, using the same procedures for establishing inputs as for the original episode.

Grid modeling applications are currently underway by or for state agencies for approximately 20 areas within the United States to support regional O_3 SIP revisions.

An immediate problem faced for almost all urban areas is that even if an adequate number of episodes exist, the episodes may not include the most adverse O_3 levels. An inherent question in using a less adverse episode to develop control strategies is how do

these strategies extrapolate to a more severe set of conditions? There is no clear answer to this question. At present, control strategies, evaluated by using grid-based models, are determined based on available episodes that have the largest amount of data, whether or not these episodes contain the highest O_3 concentration achieved. Another issue is that the form of the NAAQS for O_3 does not correspond with the output from a grid-based model. The model output does not provide a direct answer to whether an area will meet the standard in its current statistically based form.

Table 3-27 summarizes a number of recent O_3 control strategy evaluations for different areas of the United States. Some general observations can be made concerning issues that have arisen in control strategy exercises, particularly as they relate to problems associated with different areas of the country (Roth, 1992). In California, model results indicate that O_3 has been underestimated, most likely because VOC emissions from motor vehicles have been seriously underestimated. The underestimation was hidden by adjusting other model inputs within their range of uncertainty. In Atlanta, it has been estimated that approximately 60% of the VOC inventory is of biogenic origin, and the variation of anthropogenic emissions reductions required to achieve O_3 attainment within the uncertainty range of the biogenic emissions is on the order of 20%. The uncertainty range of the biogenic VOC emissions needs to be reduced to obtain tighter control strategy estimates.

The eastern United States poses special problems in regional-scale photochemical modeling. Boundary conditions typically contribute 40 to 70% of pollutant loading in many urban areas east of the Mississippi River. Regional-scale models are often either not available or not sufficiently reliable to use in estimating upwind boundary conditions. Furthermore, data are rarely available. If data are available, their use is limited to estimation of present conditions. If models are used in control strategy assessment and 40 to 70% of pollutant loading originates outside of the modeling region, major questions arise as to just how control strategies are to be determined. If uncertainties at the regional scale are significant and if regional-scale modeling is inaccurate, the limits of accuracy for urban-scale control strategy determination need to be carefully assessed.

An essential question is, given the inevitable uncertainties associated with O_3 air quality model predictions, can the effect of VOC and NO_x emissions changes on O_3 levels be unambiguously determined? The best approach to answering this question is a combination of sensitivity/uncertainty studies. Given the estimated uncertainties in model inputs and parameters for a particular application, the proposed VOC and NO_x emissions change scenarios should be examined for the full range of model inputs and parameters to determine how sensitive conclusions about the effect on O_3 levels are to the inherent uncertainties.

3.6.6 Conclusions

The 1990 CAAA (U.S. Congress, 1990) have mandated the use of photochemical grid models for demonstrating how most O_3 nonattainment areas can attain the NAAQS. Predicting O_3 is a complex problem. There are still many uncertainties in the models; nonetheless, models are useful for regulatory analysis and constitute one of the major tools for attacking the O_3 problem. These models have developed considerably in the past 10 years. However, their usefulness is constrained by having limited databases for use in model evaluation and from having to rely on hydrocarbon emissions data that may be inaccurate.

Table 3-27. Applications of Photochemical Air Quality Models to Evaluating Ozone[a]

Investigators	Region/Episode	Model Used	Strategies Evaluated
Chu et al. (1993) Chu and Cox (1993) Roselle et al. (1992) Mathur and Schere (1993)	Eastern United States; July 2-10, 1988	ROM2.2	Across-the-board NO_x/VOC reductions
Possiel et al. (1993) Possiel and Cox (1993)	Northeastern United States; July 1-12, 1988	ROM2.2	Estimate O_3 reductions per 1990 CAAA
Milford et al. (1992)	Northeastern United States; July 2-17, 1988	ROM	Analysis of effect of NO_x reductions
Rao (1987) Rao et al. (1989) Rao and Sistla (1993)	New York metropolitan area, 5 days in 1980	UAM/ROM2.1	Evaluation of 1988 SIPs and VOC/NO_x strategies
Scheffe and Morris (1990, 1991)	New York St. Louis Atlanta Dallas-Ft. Worth Philadelphia	UAM	Use of UAM for demonstrating attainment with routinely available data
Possiel et al. (1990)	Northeastern United States; July 2-17, 1988	ROM	Ozone control strategies in Northeast
Roselle and Schere (1990) Roselle et al. (1991)	Northeastern United States; July 12-18, 1980	ROM2.1	Sensitivity of O_3 in Northeast to biogenic emissions
Dunker et al. (1992a,b)	Los Angeles New York Dallas-Ft. Worth	UAM	Effects of alternate fuels and reformulated gasolines on O_3 levels
Milford et al. (1989)	South Coast Air Basin	CIT	Effects of systematic VOC and NO_x reductions
Middleton et al. (1993)	Eastern United States and southeastern Canada	RADM	2010 emissions projections

[a]See Appendix A for abbreviations and acronyms.

Primary issues and limitations associated with the use of photochemical air quality models are described below.

- High noise-to-signal ratios. Model imprecision for ozone predictions typically ranges from 25 to 40%, and inaccuracy (bias) ranges from 5 to 20%. These uncertainties are often of the same order as the percentage of reduction in the peak O_3 concentration for an area (from 160 to 120 ppb). Reasons for these inaccuracies include uncertainties in emissions inventories.
- Inadequacies of supporting databases in most geographical areas. Most areas are lacking or are deficient in data needed to estimate boundary conditions and

meteorological and air quality conditions aloft. There are few areas where speciated VOC concentrations are measured; surface NO_x data may be inaccurate. Where important data gaps exist, modeling accuracy suffers, and the prospects for reducing or eliminating the presence of compensating errors are diminished.

- Continuing need for improvements. Examples include the introduction of prognostic meteorological modeling in the mid-1980s, the discovery of underestimation of VOC emissions in the late 1980s, the inclusion of NO_x emissions from soils in 1993, and major adjustment of the emissions rates of isoprene in 1994.
- Presence of compensating errors. It appears that compensating errors have been present in many past applications, introducing the potential for bias into the estimation of the impacts of emissions control strategies.

Comparison of model predictions against ozone measurements, although necessary, is not a robust test of a model's accuracy. Ideally, one should evaluate performance against more extensive sets of species such as individual VOCs, NO_x, and NO_y. Compensating errors in input information to a model and within the model formulation can cause an O_3 model to generate correct O_3 predictions for the wrong reasons. Therefore, model evaluation indicators are needed to demonstrate the reliability of a prediction before the model can be used effectively in making control strategy decisions.

It is important to stress that, in O_3 modeling, a modeling system also is at issue, not just the air quality model itself. The modeling system includes a meteorological model, an emissions representation (where an emissions model is preferred to the traditional "inventory" approach), the air quality model, and a comprehensive supporting database. Where a problem exists, the entire modeling system must be evaluated.

Models can be used effectively in a relative sense to rank different control alternatives in terms of their effectiveness in reducing O_3 and to indicate the approximate magnitude of improvement in peak O_3 levels expected under various control strategies. To do so, there must be a sound emissions model and data and an adequate database on which to construct the modeling. Grid-based O_3 air quality modeling is superior to the available alternatives for O_3 control planning, but results can be misleading if the model is not evaluated sufficiently. The goal is to minimize the chances of incorrect use of the model.

3.7 Summary and Conclusions
3.7.1 Tropospheric Ozone Chemistry
3.7.1.1 Ozone in the Unpolluted Atmosphere
Ozone is found in the stratosphere, the "free" troposphere, and the PBL of the earth's atmosphere. In the stratosphere, O_3 is produced through cyclic reactions that are initiated by the photolysis of molecular oxygen by short-wavelength radiation from the sun and are terminated by the recombination of molecular oxygen and ground-state oxygen atoms.

In the "free" troposphere, O_3 occurs as the result of incursions from the stratosphere; upward venting from the PBL (which is the layer next to the earth, extending to altitudes of □1 to 2 km) through certain cloud processes; and photochemical formation from precursors, notably CH_4, CO, and NO_x. These processes contribute to the background O_3 in the troposphere.

Ozone is present in the PBL as the result of downward mixing from the stratosphere and free troposphere and as the result of photochemical processes occurring within the PBL. The photochemical production of O_3 and other oxidants found at the earth's surface is the result of atmospheric physical and chemical processes involving two classes of precursor pollutants, reactive VOCs and NO_x. The formation of O_3 and other oxidants from its precursors is a complex, nonlinear function of many factors, including the intensity and spectral distribution of sunlight; atmospheric mixing and related meteorological conditions; the reactivity of the mixture of organic compounds in ambient air; the concentrations of precursor compounds in ambient air; and, within reasonable concentrations ranges, the ratio between the concentrations of reactive VOCs and NO_x.

In the free troposphere and in many relatively "clean" areas of the PBL, CH_4 is the chief organic precursor to in situ photochemical production of O_3 and related oxidants. Exceptions can include clean forested or vegetated areas emitting biogenic organics. The major tropospheric removal process for CH_4 is by reaction with OH radicals. In the complex cyclic reactions that result in oxidation of CH_4, there can be a net increase in O_3 or a net loss of O_3, depending mainly on the NO concentration.

3.7.1.2 Ozone Formation in the Polluted Troposphere

The same basic processes by which CH_4 is oxidized occur in the atmospheric oxidative degradation of other, even more reactive and more complex VOCs. The only significant initiator of the photochemical formation of O_3 in the troposphere is the photolysis of NO_2, yielding NO and a ground-state oxygen atom that reacts with molecular oxygen to form O_3. The O_3 thus formed reacts with NO, yielding O_2 and NO_2. These cyclic reactions attain equilibrium in the absence of VOCs. In the presence of VOCs, however, the equilibrium is upset, resulting, from a complex series of chain reactions, in a net increase in O_3.

The key reactive species in the troposphere is the OH radical, which is responsible for initiating the oxidative degradation reactions of almost all VOCs. As in the CH_4 oxidation cycle, the conversion of NO to NO_2 during the oxidation of VOCs is accompanied by the production of O_3 and the efficient regeneration of the OH radical. The O_3 and PANs formed in polluted atmospheres increase with the NO_2/NO concentration ratio.

At night, in the absence of photolysis of reactants, the simultaneous presence of O_3 and NO_2 results in the formation of the NO_3^- radical. The reaction with NO_3^- radicals appears to constitute a major sink for alkenes, cresols, and some other compounds, although alkyl NO_3^- chemistry is not well characterized.

Most inorganic gas-phase processes, that is, the nitrogen cycle and its interrelationships with O_3 production, are well understood; the chemistry of the VOCs in ambient air, however, is not. The chemical loss processes of gas-phase VOCs, with concomitant production of O_3, include reaction with OH, NO_3, O_3, and photolysis.

The major classes of VOCs in ambient air are alkanes, alkenes (including alkenes from biogenic sources), aromatic hydrocarbons, carbonyl compounds, alcohols, and ethers. A wide range of lifetimes in the atmosphere, from minutes to years, characterize the VOCs.

The only important reaction of alkanes is with OH radicals. For alkanes having carbon-chain lengths of four or less ($\Box C_4$), the chemistry is well understood and the reaction rates are slow. For $\Box C_5$ alkanes, the situation is more complex because few reaction products have been found. Branched alkanes (e.g., isobutane) have rates of reaction that are highly dependent on structure. It is difficult to represent reactions of these VOCs satisfactorily in the

chemical mechanisms of air quality models. Stable products of alkane photooxidation are known to include carbonyl compounds, alkyl nitrates, and □-hydroxycarbonyls. Major uncertainties in the atmospheric chemistry of the alkanes concern the chemistry of alkyl nitrate formation; these uncertainties affect the amount of NO-to-NO_2 conversion occurring and, hence, the amounts of O_3 formed during photochemical degradation of the alkanes.

Alkenes react in ambient air with OH and NO_3 radicals and with O_3. All three processes are important atmospheric transformation processes, and all proceed by initial addition to the $>C=C<$ bonds. Products of alkene photooxidation include carbonyl compounds, hydroxynitrates and nitratocarbonyls, and decomposition products from the energy-rich biradicals formed in alkene-O_3 reactions. Major uncertainties in the atmospheric chemistry of the alkenes concern the products and mechanisms of their reactions with O_3, especially the radical yields (which affect the O_3 formation yields).

The only tropospherically important loss process for aromatics (benzene and the alkyl-substituted benzenes) is by reaction with the OH radical, followed by H-atom abstraction or OH radical addition. Products of aromatic hydrocarbon photooxidation include phenolic compounds, aromatic aldehydes, □-dicarbonyls (e.g., glyoxal), and unsaturated carbonyl or hydroxycarbonyl compounds. Aromatics appear to act as strong NO_x sinks under low NO_x conditions. Major uncertainties in the atmospheric chemistry of aromatic hydrocarbons are mainly with regard to reaction mechanisms and products under ambient conditions (i.e., for NO_x concentration conditions that occur in urban and rural areas). These uncertainties impact on the representation of mechanisms in models.

Tropospherically important loss processes for carbonyl compounds not containing $>C=C<$ bonds are photolysis and reaction with the OH radical; those that contain such bonds can undergo the same reactions as alkenes. Photolysis is the major loss process for HCHO (the simplest aldehyde) and acetone (the simplest ketone), as well as for the dicarbonyls. Reactions with OH radicals are calculated to be the dominant gas-phase loss process for the higher aldehydes and ketones. Products formed and the importance of photolysis are major uncertainties in the chemistry of carbonyl compounds.

Alcohols and ethers in ambient air react only with the OH radical, with the reaction proceeding primarily via H-atom abstraction from the C-H bonds in these compounds.

It should be noted that the photooxidation reactions of certain higher molecular weight VOCs can lead to the formation of significant yields of organic particulates in ambient air. The chemical processes involved in the formation of O_3 and other photochemical pollutants lead to the formation of OH radicals and oxidized VOC reaction products that are of low enough volatility to be present as organic particulate matter. Hydroxyl radicals that oxidize VOCs also react with NO_2 and SO_2 to form HNO_3 and H_2SO_4, respectively, which can become incorporated into aerosols as particulate nitrate and sulfate. Controls aimed at reducing O_3 will also impact acid and secondary aerosol formation in the atmosphere.

3.7.2 Meteorological Processess Influencing Ozone Formation and Transport
3.7.2.1 Meteorological Processes

The surface energy (radiation) budget of the earth strongly influences the dynamics of the PBL and, in combination with synoptic winds, provides the forces for the vertical fluxes of heat, mass, and momentum. The redistribution of energy through the PBL creates

thermodynamic conditions that influence vertical mixing. Energy balances require study so that more realistic simulations can be made of the structure of the PBL.

Day-to-day variability in O_3 concentrations depends heavily on day-to-day variations in meteorological conditions. For example, the concentration of an air pollutant depends significantly on the degree of mixing that occurs between the time a pollutant, or its precursors, is emitted and the arrival of the pollutant at the receptor. Inversion layers (layers in which temperature increases with height above ground level) are prominent determinants of the degree of atmospheric vertical mixing and, thus, the degree to which O_3 and other pollutants will be dispersed or accumulate. Ozone left in a layer aloft, as the result of reduced turbulence and mixing at the end of daylight hours, can be transported through the night, often to areas far removed from pollution sources. Downward mixing on the subsequent day can result in increases in local concentrations from the transported O_3.

Growing evidence indicates that the conventional use of mixing heights in modeling is an oversimplification of the complex processes by which pollutants are redistributed within urban areas. In addition, it is necessary to treat the turbulent structure of the atmosphere directly and to acknowledge the vertical variations in mixing.

Geography can significantly affect the dispersion of pollutants along the coast or shore of oceans and lakes. Temperature gradients between bodies of water and land masses influence the incidence of surface conditions. The thermodynamics of water bodies may play a significant role in some regional-scale episodes of high O_3 concentrations.

An "air mass" is a region of air, usually of multistate dimension, that exhibits similar temperature, humidity, and stability characteristics. Episodes of high O_3 concentrations in urban areas often are associated with high concentrations of O_3 in the surroundings.

The transport of O_3 and its precursors beyond the urban scale ($\square$50 km) to neighboring rural and urban areas has been well documented and was described in the 1986 EPA criteria document for O_3. Areas of O_3 accumulation are characterized by synoptic-scale subsidence of air in the free troposphere, resulting in development of an elevated inversion layer; relatively low wind speeds associated with a weak horizontal pressure gradient around a surface high pressure system; a lack of cloudiness; and high temperatures.

3.7.2.2 Meteorological Parameters

Ultraviolet radiation from the sun plays a key role in initiating the photochemical processes leading to O_3 formation and affects individual photolytic reaction steps. There is little empirical evidence in the literature, however, linking day-to-day variations in observed UV radiation levels with variations in O_3 levels.

An association between tropospheric O_3 concentrations and tropospheric temperature has been demonstrated. Plots of daily maximum O_3 concentrations versus maximum daily temperature for the summer months of 1988 to 1990 for four urban areas, for example, show an apparent upper bound on O_3 concentrations that increases with temperature. A similar qualitative relationship exists at a number of rural locations.

The relationship between wind speed and O_3 buildup varies from one part of the country to another. Research done during the SOS (in the "Atlanta intensive" field study) indicates that measurements of variations in wind speed among methods at a particular level above ground must be larger than about 3 m/s to be considered statistically significant.

3.7.2.3 Normalization of Trends

Statistical techniques (e.g., regression techniques) can be used to help identify real trends in O_3 concentrations, both intra-annual and inter-annual, by normalizing meteorological variability. In the SOS, for example, regression techniques were used successfully to forecast O_3 levels to ensure that specialized measurements were made on appropriate days.

3.7.3 Precursors

3.7.3.1 Volatile Organic Compound Emissions

Hundreds of VOCs, commonly containing from 2 to about 12 carbon atoms, are emitted by evaporative and combustion processes from a large number of source types. Total U.S. VOC emissions in 1991 were estimated at 21.0 Tg. The two largest source categories were industrial processes (10.0 Tg) and transportation (7.9 Tg). Emissions of VOCs from highway vehicles accounted for almost 75% of the transportation-related emissions; studies have shown that the majority of these VOC emissions come from about 20% of the automobiles in service, many, of which are older cars that are poorly maintained.

The accuracy of VOC emission estimates is difficult to determine, both for stationary and mobile sources. Within major area sources, deviations of emission rates from individual sources from assigned average factors can result in error for the entire area source. Evaporative emissions, which depend on temperature and other environmental factors, compound the difficulties of assigning accurate emission factors. In assigning VOC emission estimates to the mobile source category, models are used that incorporate numerous input parameters (e.g., type of fuel used, type of emission controls, age of vehicle), each of which has some degree of uncertainty.

According to recent studies, vegetation emits significant quantities of VOCs into the atmosphere, chiefly monoterpenes and isoprene, but also oxygenated VOCs. The most recent biogenic VOC emissions estimate for the United States showed annual emissions of 29.1 Tg/year. Coniferous forests are the largest vegetative contributor on a national basis, because of their extensive land coverage. Summertime biogenic emissions comprise more than half of the annual totals in all regions because of their dependence on temperature and vegetational growth. Biogenic emissions are, for those reasons, expected to be higher in the southern states than in the northern.

Uncertainties in both biogenic and anthropogenic VOC emission inventories prevent establishing the relative contributions of these two categories.

3.7.3.2 Nitrogen Oxides Emissions

Anthropogenic NO_x is associated with combustion processes. The primary pollutant emitted is NO, formed at high combustion temperatures from the nitrogen and oxygen in air and from nitrogen in combustion fuel. Emissions of NO_x in 1991 in the United States totaled 21.39 Tg. The two largest NO_x emission sources are electric power generation plants and highway vehicles. Emissions of NO_x therefore are highest in areas having a high density of electric-power-generating stations and in urban regions having high traffic densities. Between 1987 and 1991, transportation-related emissions remained essentially constant, whereas stationary source NO_x emissions increased about 10%.

Natural NO_x sources include stratospheric intrusion, oceans, lightning, soils, and wildfires. Lightning and soil emission are the only two significant natural sources of NO_x in the United States. The estimated annual lightning-produced NO_x for the continental United States is $\square$1.0 Tg, about 60% of which is generated over the southern states. Both nitrifying

and denitrifying organisms in the soil can produce NO_x, principally NO. Emission rates depend mainly on fertilization levels and soil temperature. Inventorying soil NO_x emissions is difficult because of large temporal and spatial variability, but the nationwide total has been estimated at 1.2 Tg/year, of which about 85% is emitted in spring and summer. About 60% of the total soil NO_x is emitted in the area of the country containing the central corn belt.

Combined natural sources contribute about 2.2 Tg of NO_x to the troposphere over the continental United States. Uncertainties in natural NO_x inventories are much larger than that for anthropogenic NO_x emissions. Because a large proportion of anthropogenic NO_x emissions come from distinct point sources, published annual estimates are thought to be very reliable.

3.7.3.3 Concentrations of Volatile Organic Compounds in Ambient Air

The VOCs most frequently analyzed in ambient air are NMHCs. Morning concentrations (6:00 a.m. to 9:00 a.m.) have been measured most often because of the use of morning data in EKMA and in air quality simulation models. Major field studies in 22 cities in 1984 and in 19 cities in 1985 produced NMHC measurements that showed median values ranging from 0.39 to 1.27 ppmC for 1984 and 0.38 to 1.63 ppmC in 1985. Overall median values from all urban sites were about 0.72 ppmC in 1984 and 0.60 ppmC in 1985.

Comparative data over two decades (the 1960s through the 1980s) in the Los Angeles and New York City areas showed decreases in NMHC concentrations in those areas. Concomitant compositional changes were observed over the two decades, with increases observed in the percentage of alkanes and decreases in the percentage of aromatic hydrocarbons and acetylene.

Concurrent measurements of anthropogenic and biogenic NMHCs have shown that biogenic NMHCs usually constituted much less than 10% of the total NMHCs. For example, average isoprene concentrations ranged from 0.001 to 0.020 ppmC and terpenes from 0.001 to 0.030 ppmC.

3.7.3.4 Concentrations of Nitrogen Oxides in Ambient Air

Measurements of NO_x at sites in 22 and 19 U.S. cities in 1984 and 1985, respectively, showed that median NO_x concentrations ranged from 0.02 to 0.08 ppm in most of these cities. The 6 a.m. to 9 a.m. median concentrations in many of these cities exceeded the annual average NO_x values of 0.02 to 0.03 ppm found in U.S. metropolitan areas between 1980 and 1989. Nonurban NO_x concentrations, reported as average seasonal or annual NO_x, range from <0.005 to 0.015 ppm.

Ratios of 6 a.m. to 9 a.m. NMOC to NO_x are higher in southeastern and southwestern U.S. cities than in northeastern and midwestern U.S. cities, according to data from EPA's multi-city studies conducted in 1984 and 1985. Median ratios ranged from 9.1 to 37.7 in 1984; in 1985, median ratios ranged from 6.5 to 53.2 in the cities studied. Rural NMOC/NO_x ratios tend to be higher than urban ratios. Morning (6 a.m. to 9 a.m.) NMOC/NO_x ratios are used in the EKMA-type of trajectory model. Trends from 1976 to 1990 show decreases in these ratios in the South Coast Air Basin of California. The correlation of NMOC/NO_x ratios with maximum 1-h O_3 concentrations, however, was weak in a recent analysis.

3.7.3.5 Ratios of Concentrations of Nonmethane Organic Compounds to Nitrogen Oxides

The ratios of NMOC/NO_x vary substantially between cities and within a given city. With certain exceptions, urban NMOC/NO_x ratios have been in the range of 10 and below. In contrast, ratios of NMOC/NO_x in rural areas tend to equal or exceed 20. Discrepancies have been found between ambient NMOC/NO_x ratios and emission inventory NMOC/NO_x ratios, with ambient ratios of NMOC/NO_x significantly exceeding emission ratios of NMOC/NO_x.

Trends in ratios of NMOC/NO_x have shown downward trends to well below 10 during the 1980s, both for the South Coast Air Basin and for cities in the eastern United States. Based on these low ratios, hydrocarbon control should be more effective than NO_x control within a number of cities.

3.7.3.6 Source Apportionment and Reconciliation

Source apportionment (now regarded as synonymous with receptor modeling) refers to determining the quantitative contributions of various sources of VOCs to ambient air pollutant concentrations. Source reconciliation refers to the comparison of measured ambient VOC concentrations with emissions inventory estimates of VOC source emission rates for the purpose of validating the inventories.

Early studies in Los Angeles employing a "mass balance" approach to receptor modeling showed the following estimated contributions of respective sources to ambient air concentrations of NMOCs through C_{10}: automotive exhaust, 53%; whole gasoline evaporation, 12%; gasoline headspace vapor, 10%; commercial natural gas, 5%; geogenic natural gas, 19%; and liquefied natural gas, 1%. Recent studies in eight U.S. cities showed that vehicle exhaust was the dominant contributor to ambient VOCs (except in Beaumont, where 14% was reported). Estimates of the contributions of gasoline evaporation differ in methodology; the more appropriate methods used result in estimates of large whole gasoline contributions (i.e., equal to vehicle exhaust in one study and 20% of vehicle exhaust in a second study).

The chemical mass balance approach used for estimating anthropogenic VOC contributions to ambient air cannot be used for receptor modeling of biogenic sources. A modified approach, applied to 1990 data from a downtown site in Atlanta, indicated a lower limit of 2% (24-h average) for the biogenic percentage of total ambient VOCs at that location (isoprene was used as the biogenic indicator species). The percentage varies during the 24-h period because of the diurnal (e.g., temperature, light intensity) dependence of isoprene concentrations.

Source reconciliation data have shown disparities between emission inventory estimates and receptor-estimated contributions. For biogenics, emission estimates are greater than receptor-estimated contributions. The reverse has been true for natural gas contributions estimated for Los Angeles, Columbus, and Atlanta and for refinery emissions in Chicago.

3.7.4 Analytical Methods for Oxidants and Their Precursors

3.7.4.1 Oxidants

Current methods used to measure O_3 are CL, UV absorption spectrometry, and newly developed spectroscopic and chemical approaches, including chemical approaches applied to passive sampling devices for O_3.

The CL method, designated as the reference method by EPA, involves the direct gas-phase reaction of O_3 with an alkene (C_2H_4) to produce electronically excited products, which decay with the emission of light. Detection limits of 0.005 ppm and a response time of less than 30 s are typical of currently available commercial instruments. A positive interference from atmospheric water vapor was reported in the 1970s and has recently been confirmed. Proper calibration can minimize this source of error.

Commercial UV photometers for measuring O_3 have detection limits of about 0.005 ppm, long-term precision within about $\pm 5\%$, and a response time of <1 min. Ozone has a fairly strong absorption band with a maximum near 254 nm; its molar absorption coefficient at that wavelength is well known. Because the measurement is absolute, UV photometry also is used to calibrate other O_3 methods.

A potential disadvantage of UV photometry is that atmospheric constituents that absorb 254-nm radiation (and that are removed fully or partially by the MnO_2 scrubber used in UV O_3 photometers) will be positive interferences in O_3 measurements. Interferences have been reported in two recent studies but assessment of the potential importance of such interferences (e.g., toluene, styrene, cresols, nitrocresols) is hindered by lack of absorption spectra data in the 250-nm range and by lack of ambient measurements of most of the aromatic photochemical reaction products. An interference from water also appears to occur from condensation of moisture in sampling level. Results from collocated UV and CL instruments indicated positive biases in the UV data of 20 to 40 ppb on hot, humid days.

Differential optical absorption spectrometry has been used to measure ambient O_3, but further intercomparisons with other methods and interference tests are recommended. Passive sampling devices permit acquisition of personal human exposure data and of O_3 monitoring data in areas where the use of instrumental methods is not feasible. Three PSDs are commercially available; all employ solid absorbents that react with O_3.

Calibration of O_3 measurement methods (other than PSDs) is done by UV spectrometry or by GPT of O_3 with NO. Ultraviolet photometry is the reference calibration method approved by EPA. Ozone is unstable and must be generated in situ at time of use to produce calibration mixtures.

Two methods generally have been employed to measure atmospheric PAN and its higher homologues: IR and GC using an ECD. A third method, less often used, couples GC with a molybdenum converter that reduces PAN to NO in the gas phase and subsequently measures the NO with a CL analyzer. Peroxyacetyl nitrate and the higher PANs are normally measured by GC-ECD. Detection limits have been extended to 1 to 5 ppt, using cryogenic enrichment of samples and specified desorption procedures that limit losses associated with cryosampling. Because PAN is unstable (explosive, and subject to surface-related decomposition), the preparation of reliable calibration standards is difficult. Methods devised to generate calibration standards include photolysis of static concentrations of gases, nitration of peracetic acid in single hydrocarbons, and analysis of PAN as NO under specified conditions of the dissociation of PAN into its precursors.

Early measurements of 10 to 80 ppb H_2O_2 reported in the 1970s have been found to be in error because of artifact formation of H_2O_2 from reactions of absorbed gaseous O_3. Modeling results also indicate that lower levels of H_2O_2, on the order of 1 ppb, occur in the atmosphere.

In situ measurement methods for H_2O_2 include FTIR and TDLAS. The FTIR method is specific for H_2O_2 but has a high detection level of $\Box$50 ppb (using a 1-km path

length). The TDLAS method also is specific and has a detection level of 0.1 ppb over averaging times of several minutes. Four frequently used wet chemical methods for measurement of H_2O_2 are available. All involve the oxidation of a substrate followed by instrumental detection and quantification of the resulting CL or fluorescence. Detection limits are comparable to those of FTIR and TDLAS, but interferences are common and must be obviated or minimized with specified procedures.

Calibration of methods for gaseous H_2O_2 measurement requires the immediate use of standard mixtures prepared by one of several wet chemical methods.

3.7.4.2 Volatile Organic Compounds

Increased monitoring of VOCs is required under Title I, Section 182, of the CAAA of 1990 because of the role of VOCs as precursors to the formation of O_3 and other photochemical oxidants. Volatile organic compounds are those gaseous organic compounds that have a vapor pressure greater than 0.15 mm and, generally, have a carbon content ranging from C_1 through C_{12}.

Traditionally, NMHCs have been measured by methods that employ a FID as the sensing element that measures a change in ion intensity resulting from the combustion of air containing organic compounds. The method recommended by EPA for total NMOC measurement involves the cryogenic preconcentration of nonmethane organic compounds and the measurement of the revolatilized NMOCs using FID. The main technique for speciated NMOC/NMHC measurements is cryogenic preconcentration followed by GC-FID. Systems for sampling and analysis of VOCs have been developed that require no liquid cryogen for operation, yet provide sufficient resolution of species.

Stainless steel canisters have become the containers of choice for collection of whole-air samples for NMHC/NMOC data. Calibration procedures for NMOC instrumentation require the generation, by static or dynamic systems, of dilute mixtures at concentrations expected to occur in ambient air.

Preferred methods for measuring carbonyl species (aldehydes and ketones) in ambient air are spectroscopic methods, on-line colorimetric methods, and HPLC method employing DNPH derivatization in a silica gel cartridge. The most common method in current use for measuring aldehydes in ambient air is the HPLC-DNPH method. Use of an O_3 scrubber has been recommended to prevent interference in this method by O_3 in ambient air. Carbonyl species are reactive, making preparation of stable calibration mixtures difficult; but several methods are available.

Impetus for the development of methods for measuring the more reactive oxygen- and nitrogen-containing organic compounds has come from their roles as precursors or products of photochemical oxidation and also from the inclusion of many of these compounds on the list of hazardous air pollutants in the 1990 CAAA. Measurement of these PVOCs is difficult because of their reactivity and water solubility. Methods are still in development.

3.7.4.3 Oxides of Nitrogen

Nitric oxide and NO_2 comprise the NO_x involved as precursors to O_3 and other photochemical oxidants.

The most common method of NO measurement is the gas-phase CL reaction with O_3. The CL method is essentially specific for NO. Commercial NO monitors have detection limits of a few parts per billion by volume in ambient air. Commercial NO analyzers may not

have sensitivity sufficient for surface measurements in rural or remote areas or for airborne measurements. Direct spectroscopic methods for NO exist that have very high sensitivity and selectivity for NO. Major drawbacks of these methods are their complexity, size, and cost, which restrict these methods to research applications. No PSDs exist for measurement of NO.

Chemiluminescence analyzers are the method of choice for NO_2 measurement, even though they do not measure NO_2 directly. Minimum detection levels for NO_2 have been reported to be 5 to 13 ppb, but more recent evaluations have indicated detection limits of 0.5 to 1 ppbv. Reduction of NO_2 to NO is required for measurement. In practice, selective measurement of NO_x by this approach has proved difficult. Commercial instruments that use heated catalytic converters to reduce NO_2 to NO measure not NO and NO_x, but more nearly NO and total NO_y. Thus, the NO_2 value inferred from such measurements may be significantly in error, which may in turn affect the results of modeling of ambient O_3.

Several spectroscopic approaches to NO_2 detection have been developed. As noted above for NO, however, these methods have major drawbacks that include their complexity, size, and cost, which, at present, outweigh the advantages of their sensitivity and selectivity. Passive samplers for NO_2 exist but are still in the developmental stage for ambient air monitoring.

Calibration of methods for NO measurement is done using standard cylinders of NO in nitrogen. Calibration of methods for NO_2 measurement include the use of cylinders of NO_2 in nitrogen or air, the use of permeation tubes, and gas-phase titration.

3.7.5 Ozone Air Quality Models
3.7.5.1 Definitions, Descriptions, and Uses

Photochemical air quality models are used to predict how O_3 concentrations change in response to prescribed changes in source emissions of NO_x and VOCs. They are mathematical descriptions of the atmospheric transport, diffusion, removal, and chemical reactions of pollutants. They operate on sets of input data that characterize the emissions, topography, and meteorology of a region and produce outputs that describe air quality in that region.

Two kinds of photochemical models are recommended in guidelines issued by EPA: (1) the grid-based UAM is recommended for modeling O_3 over urban areas, and (2) EKMA is identified as an acceptable approach under certain circumstances. The 1990 CAAA mandate the use of three-dimensional (grid-based) air quality models such as UAM in developing SIPs for areas designated as extreme, severe, serious, or multistate moderate.

In grid-based air quality models, the region to be modeled (the modeling domain) is subdivided into a three-dimensional array of grid cells. Pertinent atmospheric processes and chemical reactions are represented for each cell.

In trajectory models, such as EKMA, a hypothetical air parcel moves through the area of interest along a path calculated from wind trajectories. Emissions are injected into the air parcel and undergo vertical mixing and chemical transformations. Trajectory models provide a dynamic description of atmospheric source-receptor relationships that is simpler and less expensive to derive than that obtained from grid models, but meterological processes are highly simplified in trajectory models

The EKMA-based method for determining O_3 control strategies has some limitations, the most serious of which is that predicted emissions reductions are critically dependent on the initial NMHC/NO_x ratio used in the calculations. This ratio cannot be

determined with any certainty because it is expected to be quite variable in time and space in an urban area. Grid-based models have their limitations as well. These are pointed out subsequently.

3.7.5.2 Model Components

Spatial and temporal characteristics of VOC and NO_x emissions are major inputs to a photochemical air quality model. Greater accuracy in emissions inventories is needed, for biogenics and for both mobile and stationary source components. Grid-based air quality models also require as input the three-dimensional wind field for the photochemical episode being simulated. This input is supplied by "meteorological modules" which fall into one of four categories: (1) objective analysis procedures; (2) diagnostic methods; (3) dynamic, or prognostic, methods; and (4) hybrid methods that embody elements from both diagnostic and prognostic approaches. Prognostic models are believed to provide a dynamically consistent, physically realistic, three-dimensional representation of the wind and other meteorological variables at scales of motion not resolvable by available observations. Outputs of prognostic models do not always agree with observational data, but methods have been devised to mitigate these problems.

A chemical kinetic mechanism (a set of chemical reactions), representing the important reactions that occur in the atmosphere, is used in an air quality model to estimate the net rate of formation of each pollutant simulated as a function of time. Chemical mechanisms that explicitly treat each individual VOC component of ambient air are too lengthy to be incorporated into three-dimensional atmospheric models. " Lumped" mechanisms are therefore used. The chemical mechanisms used in existing photochemical O_3 models contain uncertainties that may limit the accuracy of their predictions. Because of different approaches to "lumping" of reactions, models can produce somewhat different results under similar conditions. Both the UAM (UAM-IV) and EPA's ROM use the CMB-IV. The CBM-IV and the SAPRC and RADM mechanisms are considered to represent the state of the science.

Dry deposition, the removal of chemical species from the atmosphere by interaction with ground-level surfaces, is an important removal process for O_3 on both urban and regional scales; and is included in all urban- and regional-scale models. Wet deposition (the removal of gases and particles from the atmosphere by precipitation events) generally is not included in urban-scale photochemical models, because O_3 episodes do not occur during periods of significant clouds or rain.

Concentration fields of all species computed by the model must be specified at the beginning of the simulation; these concentration fields are called the initial conditions. These initial conditions are determined mainly with ambient measurements, either from routinely collected data or from special studies, but interpolation can be used to distribute the surface ambient measurements.

3.7.5.3 Evaluation of Model Performance

Air quality models are evaluated by comparing their predictions with ambient observations. An adequate model should give accurate predictions of current peak O_3 concentrations and temporal and spatial O_3 patterns. It also should respond accurately to changes in VOC and NO_x emissions, to differences in VOC reactivity, and to spatial and temporal changes in emissions patterns for future years. Likewise, multispecies comparisons could be the key in discriminating among alternative modeling approaches that provide similar

predictions of O_3 concentrations. Adequate model performance for several reactive species increases the assurance that correct O_3 predictions are not a result of chance or fortuitous cancellation of errors introduced by various assumptions.

If only a routine database is available for modeling O_3 in an urban area, then several concerns require attention relative to model performance evaluation: air quality aloft, boundary conditions, ambient VOC data, and meteorological data aloft. If any of these four areas is missing from the database, the performance evaluation and subsequent model application must be adequately planned to minimize the possibility of compensatory errors.

3.7.5.4 Use of Ozone Air Quality Model for Evaluating Control Strategies

Photochemical air quality models are used for control strategy evaluation by first demonstrating that a past episode, or episodes, can be simulated adequately and then reducing hydrocarbon or NO_x emissions, or both, in the model inputs and assessing the effects of these reductions on O_3 in the region. The adequacy of control strategies based on grid-based models depends, in part, on the nature of input data for simulations and model validation, on input emissions inventory data, and on the relationship between model output and the current form of the NAAQS for O_3.

Grid-based models that have been widely used to evaluate control strategies for O_3 or acid deposition, or both, are the UAM, the CIT model, the ROM, the ADOM, and the RADM.

3.7.5.5 Conclusions

Urban air quality models are becoming readily available for application and have been applied in recent years in several urban areas. Significant progress also has been made in the development of regional models and in the integration of state-of-the-art prognostic meteorological models as drivers.

There are still many uncertainties in photochemical air quality modeling. Prime among these are emission inventories. However, models are essential for regulatory analysis and constitute one of the major tools for attacking the O_3 problem. Grid-based O_3 air quality modeling is superior to the available alternatives for O_3 control planning, but the chances of its incorrect use must be minimized.

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